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FY04-Q4 REPORT: LX-17 MODELING

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Disclaimer

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1.0 – LX-17 Ratchet Growth

TATB containing explosives tend to permanently expand as their temperatures are increased or thermally cycled, a phenomenon known as "ratchet-growth." Mesoscale simulations using dissipative particle dynamics (DPD) have been carried out in order to study the geometric packing effects of TATB pressed powders under stress conditions. Further, our mesoscale simulations of polycrystalline TATB pressed powders have been used to predict hot-spot changes as a function of temperature (thermal cycling) and confinement. Our DPD simulations showed irreversible permanent growth in the mesoscale pressed powders *only* when crystal fracture induced by the anisotropic thermal expansion of TATB was incorporated into the model.

1.1.1 Goals of the TATB Modeling Effort

The properties of the solid energetic compound 1,3,5-triamino-2,4,6-trinitrobenzene (TATB) is an important high-energy material, and has generated considerable interest since its first synthesis,[1] due to its nonlinear optical properties,[2-4] as well as its extraordinary stability under thermal, impact or shock initiation conditions.[5,6] Additionally, TATB containing explosives are known to permanently grow as they are thermally cycled.

During the 2004 fiscal, our focus has been on determining the overall structural evolution and void size distributions in TATB pressed powders under different stress conditions. Further, it is desired to determine if the structural changes manifest in TATB pressed powders are controlled by the orientation and anisotropic expansion of individual grains (i.e., cumulative damage events which may lead to expansion into voids structures created by microcracking which occurs during thermal cycling). The results of our findings are intended to be used as direct input into the age aware hydro model which is be developed concurrently.

1.1.2 Mesoscale TATB Simulations

In previous years it was determined that the irreversible growth in TATB pressed powders is associated crystal fraction as well as the anisotropic thermal expansion in TATB. Here we use our newly developed mesoscale powder TATB model to investigate the non-reversible growth under various stress conditions, coupled with the DPD simulation methodology. The DPD simulations were carried out to study TATB's structural evolution and geometric packing Our DPD model for TATB consists of a ternary distribution of TATB crystallites composed of 10-5x5x5, 3-6x6x6, and 2-8x8x8 DPD molecular clusters or "beads," where each spherical bead, having a diameter of 0.02

μm, represents a cluster of TATB molecules (~$4.5x10^9$ TATB molecules), which gives an average crystallite particle size distribution of ~5.6 μm. An illustration of our mesoscale DPD TATB model is shown in Figure 1.

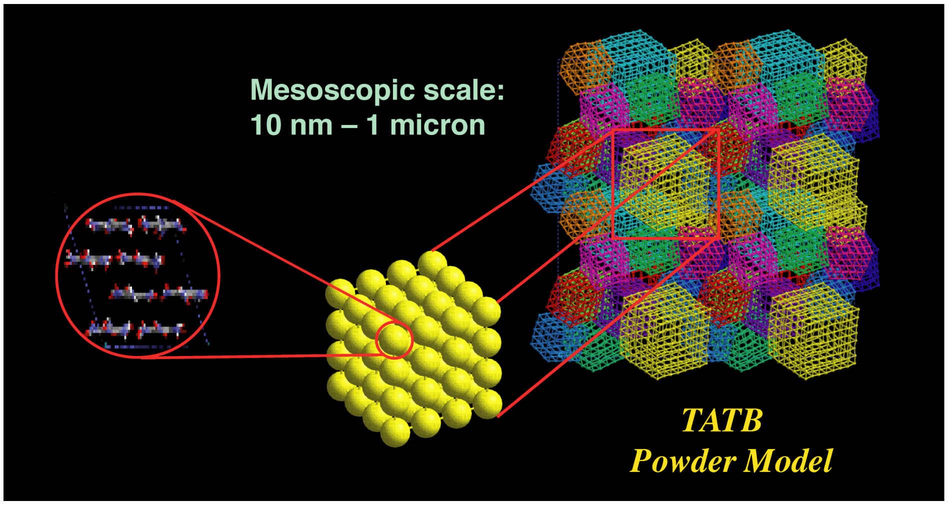

Figure 1: Illustration of the mesoscale TATB powder model consisting of a ternary distribution of TATB crystallites. Each spherical bead in the crystallite lattice represents a molecular cluster of ~$4.5X10^9$ TATB molecules.

1.1.3 Ratchet-Growth: Void Volume Evolution Under Externally Imposed Stress and Confinement

As mentioned, our mesoscale TATB model manifests non-reversible growth *only* when crystal fracture and anisotropic thermal expansion were incorporated into the model, thus, suggesting that crystal fracture induced by the anisotropic volume expansion of TATB is the root cause for the permanent growth seen in TATB containing explosives. Our results, with no external impedes (e.g., external stresses) were found to compare well to experiment. Based on the success of our TATB powder model, we have performed DPD simulations where external stresses were imposed. First, simulations were performed with an applied isotropic hydrostatic pressure. The underlying irreversible volume increase in the TATB pressed powders under isotropic hydrostatic loading was found to be due to cumulative damage events which may lead to expansion into voids structures created by microcracking which occurs during thermal cycling.

Figure 2 shows the evolution of our mesoscale TATB model as a function thermal cycling both at ambient and isotropic hydrostatic pressures. The systems were temperature cycled in *NPT* runs starting at –140 ^{o}C and incrementally increasing the temperature in steps of 43.5 ^{o}C to a maximum temperature of 83 ^{o}C, and then incrementally cooled using the same approach. The systems were allowed to dynamically evolve at each temperature for a total of either 0.25 s before the temperature was incremented. The dimensional change as a function of thermal cycling is shown in Figure 2. As is apparent the overall irreversible volumetric growth is significantly reduced under isotropic hydrostatic loading.

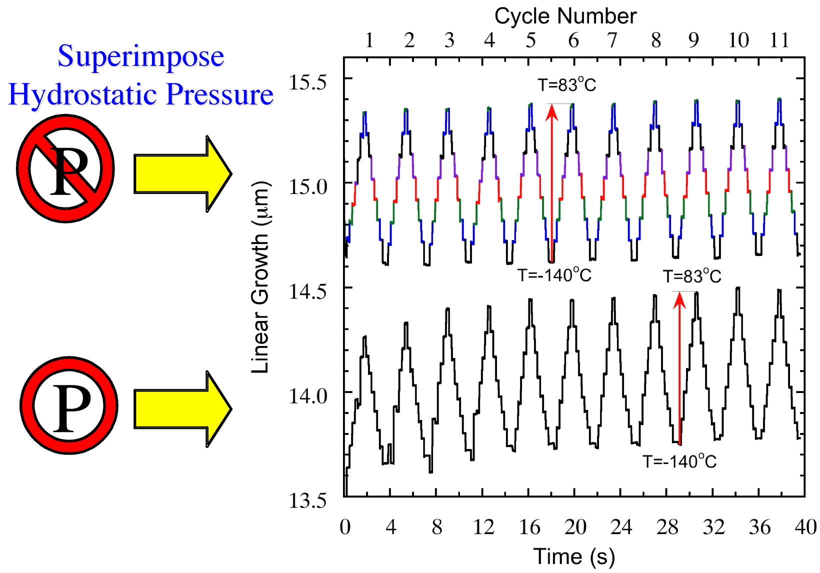

Figure 2: Dimensional change of the mesoscale TATB pressed powder model as a function of time and isotropic hydrostatic pressure showing identifiable hysteresis. The temperature is incrementally changed from -140 ^{o}C to 83 ^{o}C over time.

The evolution of the void volume, as illustrated in Figure 3, as a function of thermal cycling with no external hydrostatic pressure shows that the overall void volume as well as the average void size slightly increases as a function of thermal cycle as shown in Figure 4*a*. Figure 4 shows that the void volume size distribution tends toward larges voids, at the expense of smaller voids upon thermal cycling.

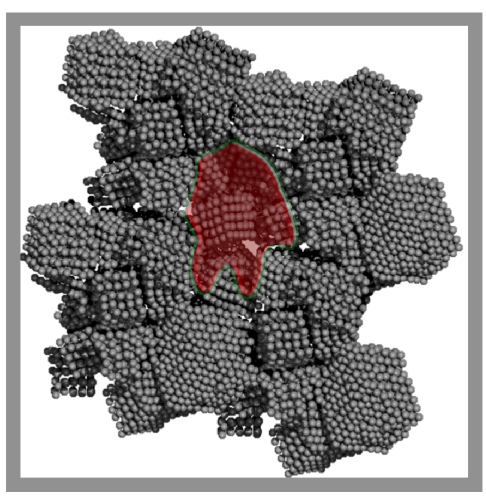

Figure 3: Illustration of void volume within the TATB pressed powder model.

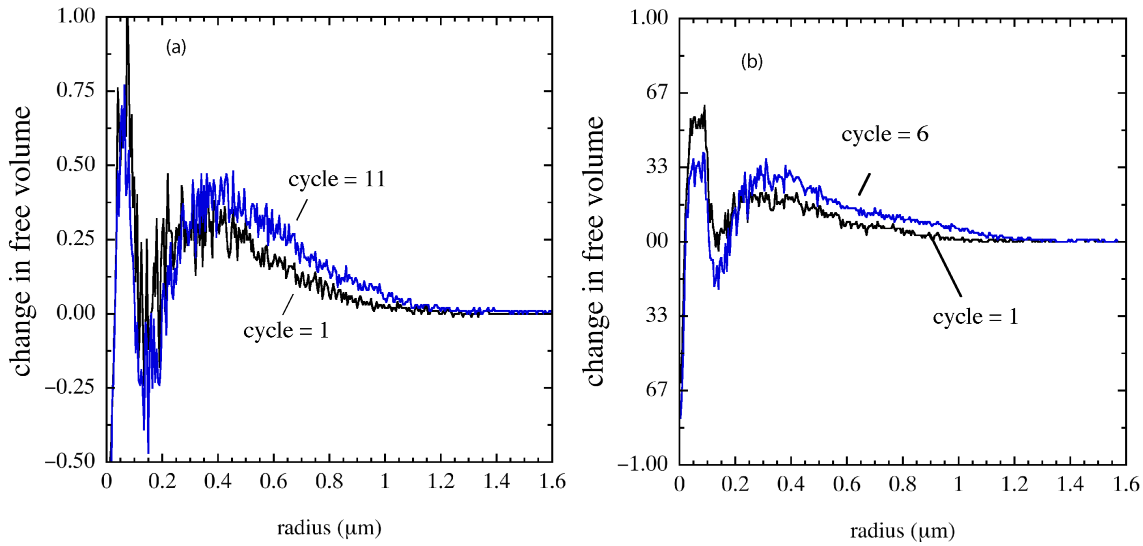

Figure 4: Void volume size distribution as a function of thermal cycle. Panel (*a*) shows results for simulations performed with out hydrostatic pressure, where panel (*b*) shows results where the TATB powder is confined in one dimension.

Similar behavior is manifest when the TATB pressed powder ensemble is confined in one-dimension (Figure 4*b*).

1.1.4 Ratchet-Growth: Crystal Size Evolution Under Externally Imposed Stress and Confinement

To further understand the overall structural changes in the TATB powder under both one-dimensional confinement and constant isotropic hydrostatic pressures we have attempted to determined the morphological changes in the TATB crystallites, as well as track the TATB crystallite size distributions as a function of thermal cycling. We first focus on the morphological changes in TATB as a function of thermal cycling, namely, do the individual beads which make up the crystallites remain associated with the initial crystallite, or do bead effectively trade places with one another (i.e., do clusters of beads swap members). Further, it is of interest to determine whether the individual crystallites are expanding extensively (i.e., are the beads within the crystallites translating significantly). The simulations show effectively no change in the overall particle size upon thermal cycling (see Figure 5).

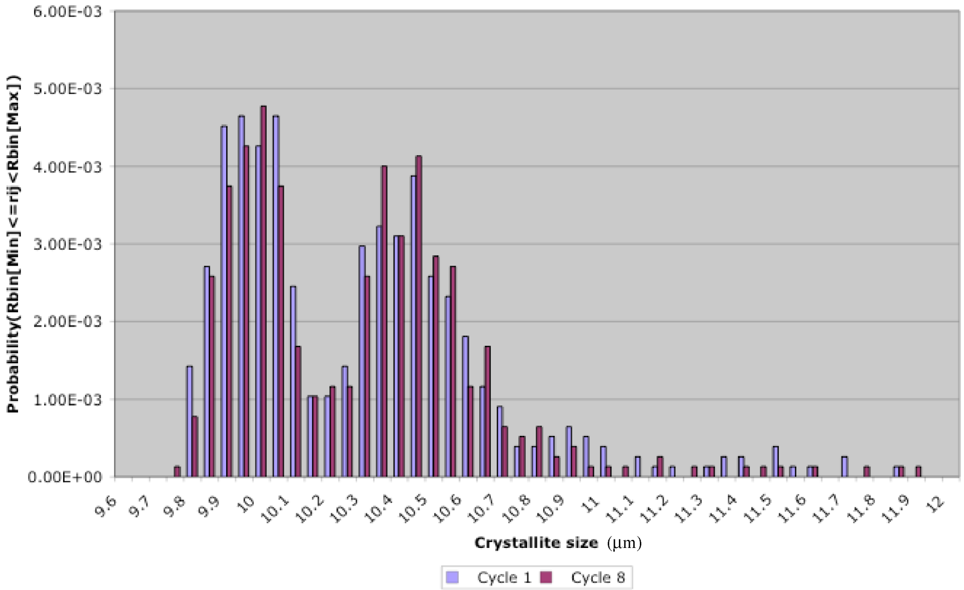

Figure 5: Crystal particle size distribution as a function of thermal cycling (-140 to 83 ^{o}C) under one-dimensional confinement.

1.1.5 Conclusions from Mesoscale TATB Thermal Cycling Modeling Studies

In conclusion, our simulations of pressed TATB powders under hydrostatic pressure and one-dimensional confinement show an overall increase in the available void volume as a function of thermal cycling, but show no indication of changes in the crystallite particle size. However, the characteristics associated with the increase in void volume and irreversible volume growth seems to be intricately associated with TATB's anisotropic volume expansion.